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PRE-IRRADIATION ANNEALING AND FADING CHARACTERISTICS FOR CLEAR POLYMETHYLMETHACRYLATE PERSONNEL DOSIMETER

M. B. H. AL-BEDRI[1] & I. J. ABDUL GHANI[2]

[1]Department of Geology, Faculty of Science, University of Jordan, Amman, Jordan

[2]Department of Physics, Faculty of Education for Pure Sciences, University of Baghdad, A'Adamiya, Baghdad, Iraq

ABSTRACT

The pre-irradiation annealing technique (Pre-IAT) at annealing temperature of 60°C and storage time of 48 h (60°C /48 h) of clear Polymethylmethacrylate (PMMA) personnel dosimeter have been studied. Pre-irradiation and post-irradiation fading characteristics of PMMA were measured at storage times up to 90 days and at six different temperatures (0, 10, 25, 40, 50 and 60^{o}C). PMMA personnel dosimeters stability after irradiated with 0.5 Gy test absorbed dose of β-particles have been investigated. The response of personnel dosimeters during pre- and post-irradiation storage at 60^{o}C/48 h shows good stability for at least 90 days. PMMA samples were irradiated with β-particles from ^{90}Sr beta source showed a decrease in the mean molecular weight with increasing absorbed dose range from 20 mGy – 10 Gy, which is attributed to chain scission of PMMA.

KEYWORDS: PMMA Personnel Dosimeter, Pre-Irradiation Annealing Technique, Pre-and Post-Irradiation Fading, β-Particles Absorbed Dose, Molecular Weight, Degradation

INTRODUCTION

Some of the polymers that are used as a dosimeter are: Polymethylmethacrylate clear-PMMA e.g. Radix RN15 (R) [1], Radix W [2] and dyed-PMMA dosimeter e.g. [Red 4034 Harwell dosimeters [3], Blue PMMA [4], Amber 3042 and Gammachrome YR [5], and undyed Polyvinylchloride (PVC) [6,7], Poly(ethylene terephthalate- PET) [8], and Poly(lactic acid) [9,10]. Several kinds of dyed and undyed polymers are widely used to control and supervise any application of ionizing radiation in industrial processing of materials and products [1, 2, and 11]. Various kinds of polymers irradiated with UV [12], X-rays [13], gamma-rays [3, 5, 9, 11], electrons [2, 7–9] and neutrons [3, 6] have been studied using different techniques. Many techniques have been developed to measure the molecular weight of the polymers such as: Osmometry [14], Viscometry [6, 8, 10, 15], Light Scattering (LS) [16], Low-Angle Laser Light Scattering Photometry (LALLS) [17], UV Spectrophotometry [2, 8, 9, 12], and Gel Permeation Chromatography (GPC) [18]. Viscosity technique is very popular because it is experimentally simple, not expensive, reproducible and reliable technique with optimum efficiency used for the evaluation the characteristics of polymer samples [12, 19, and 20].

PMMA is classified as routine dosimeter systems for measuring absorbed doses in materials irradiated by photons or electrons in terms of absorbed dose in water [6, 8, 10, 15, 20 and 21]. The PMMA dosimetry systems are commonly used in industrial radiation processing, for example, in the sterilization of disposable medical devices and the quality assurance of the irradiation of foods [1, 2, 11 and 22]. In these applications, absorbed doses fall mostly within 0.1 kGy to 100 kGy working range of the family of PMMA dosimeters [20].

The intrinsic viscosity [η] is an outstanding parameter determined by the solution viscosity and is a measure of the volume of the polymer in a given solvent. Intrinsic viscosity measurements are used to determine the average molecular weight of the polymer samples by applying Mark-Houwink relation [23]. The materials utilized in radiation dosimetry must have a high sensitivity to ionizing radiation within the dose ranges required for the specific application [11], must also show stability, reliability, minimum post-irradiation fading and ease of use for all applications. The advantages of using PMMA dosimeters [2, 5, 11, 12, 24] are: rigid polymer, cheap, stable, reliable, ease of use and transported easily, while the disadvantages of these dosimeters that their dose response are affected by the temperature [2, 21, 25, 26].

Till today, to our knowledge, no experimental data are available in the literature on the study of the characteristics of PMMA personnel dosimeters for beta-particles [2]. In this study, the effects of pre-irradiation annealing technique (Pre-IAT) at annealing temperature of 60^{o}C and annealing period of 48 h (60^{o}C/48h) on the molecular weight of clear PMMA have been investigated in details. The characteristics of pre- and post-irradiation fading on clear PMMA dosimeter at different annealing storage temperatures (0, 10, 25, 40, 50 and 60^{o}C) and storage time up to 90 days have been examined. The stability of PMMA dosimeter post irradiation with 0.5 Gy test absorbed dose from ^{90}Sr beta source have been investigated. The changes in the mean molecular weights of the PMMA samples irradiated with beta particles (20 mGy – 10 Gy) from ^{90}Sr source have been studied.

EXPERIMENTAL

Preparation of Solutions

The clear PMMA samples were obtained from ICI, England in the form of sheets of 1 mm thickness. 0.5 g of PMMA sheet was dissolved in 100 ml of pure chloroform. Dilute solutions in the concentration range from 0.10 to 0.50 g/dl were prepared under stirring at 25^{o}C. The temperature was thermostatically controlled to within ± 0.1^{o}C. Viscometry experiments were carried out by using an automatic capillary viscometer type Viscomatic-152000 Fica, France. The flow time of pure chloroform was measured automatically at 25 ± 0.1^{o}C. A 10 ml volume of PMMA solution was pipette into the viscometer for each determination. The flow time was measured automatically for a fixed volume of PMMA solution which flew through a capillary tube of a calibrated glass viscometer under an accurate automatic reproducible head and at a closely controlled temperature of 25 ± 0.1^{o}C. The flow time used in all subsequent calculations of reduced viscosity (η_r) was the average of at least ten readings, which agreed to within ±1 ms. The viscosity of PMMA solution was calculated from the measured flow time at 25 ± 0.1^{o}C, while the intrinsic viscosity [η] values were obtained by extrapolating the reduced viscosity ($\eta_r = \eta_{sp/C}$) to zero concentration [6, 27]. Determination of intrinsic viscosity [η] was used to calculate the average molecular weight of PMMA solution by applying Mark-Houwink relation [23]. The semi-empirical Mark-Houwink equation gives a relation between intrinsic viscosity [η] and the average molecular weight (M):

$$[\eta] = KM^a \tag{1}$$

where a and K are constants depends on the particular polymer, solvent and temperature during measurements. The values of K = 4.8x10^{-3} ml/g and a = 0.80 for clear PMMA samples in pure chloroform at 25^{o}C [28].

Irradiation of PMMA Sheets

The main effects of ionizing radiation on polymers can be divided into two main sections; firstly main-chain scission (degradation) [13, 18, 19], which results in a reduction in a molecular weight, and secondly cross linking [7],

which results in an increase in a molecular weight of the polymers. Irradiated polymers with ionizing radiation create free radicals which will often chemically react in various ways, sometimes at slow reaction rates and can recombine forming the crosslinking [8]. In many polymers both processes (chain scission and cross linking) occur at the same time [6, 8, 10], where one may be predominate over the other, depending upon the type of polymer and the amount of the absorbed dose. However in certain cases the main-chain scission predominates the crosslinking and such polymers are known as degrading polymers as polymethylmethacrylate (PMMA) [11, 13, 18, 19]. The degree of degradation in PMMA is normally proportional to the amount of absorbed dose received by polymer and is generally reproducible [6, 29]. Such changes can be used as a measure of radiation doses in the polymers [2, 20].

The PMMA samples in the form of sheets were irradiated in air at room temperature (25^{o}C) and a distance of 40 cm from calibrated ^{90}Sr beta source of a dose rate 24 mGy/min obtained from the Radiochemical center, Amersham, England. Parallel-plate calibrated ionization chamber was attached to the PMMA dosimeter to evaluate the real doses received by PMMA sheets. The doses were measured at the same time of irradiation using parallel-plate free-air calibrated ionization chamber. The samples were irradiated with doses ranging from 20 mGy to 10 Gy. Agreement within a few percent was obtained for the two methods.

RESULTS AND DISCUSSIONS

Pre-Irradiation Annealing Technique (Pre-IAT)

Usually, the dose response of the polymers are affected by the environmental circumstances such us temperature [2, 6, 21, 25]. The first matter to be investigated in this study was the application of pre-irradiation annealing technique (Pre-IAT) to the clear PMMA personnel dosimeters in order to eliminate the effects of variation of temperatures during winter and summer seasons without causing any damage to this material. This technique was done to both pre- and post-irradiation (Post-I) processes in order to increase the sensitivity of the polymers. The highest storage temperature of 60^{o}C represents the unusual environmental conditions in summer time in the Middle East countries. One hundred sheets of pre-irradiated PMMA samples were annealed in a stabilized oven at 60^{o}C and were kept for different storage time of 0, 6, 12, 18, 24, 30, 40, 48, 72 and 96 hours. The sensitivity of unirradiated PMMA personnel dosimeters were determined by measuring the mean molecular weight.

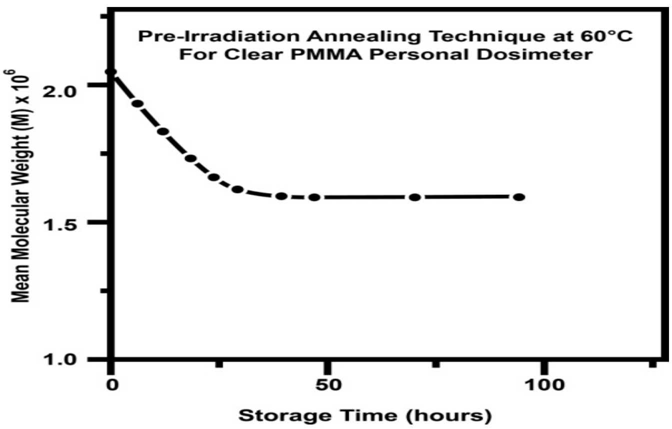

Figure 1: Variation of the Mean Molecular Weight of Un-Irradiated Clear PMMA Personnel Dosimeters as a Function of Storage Time after Applying Pre-IAT at 60^{o}c

Figure 1 illustrates the decrease of the mean molecular weight with increasing storage time up to 40 h and then reaches stability at 48 h up to 96 h. Such a decrease in the mean molecular weight of PMMA before irradiation is known as pre-irradiation fading [30]. Similar fading effects have also been previously observed in polyvinyl chloride irradiated with fast neutrons [6]. The sensitivity of clear PMMA dosimeters decreases by 22.7% in 48 hours. From the analysis of the experimental data (Figure 1), it was found that the optimum pre-irradiation annealing condition was 60^{o}C for storage period of 48 h (60^{o}C/48 h). This period of 48 h and annealed temperature of 60^{o}C were found to be sufficient for the molecular weight sensitivity of the PMMA samples to reach stable state and to be resistant to the variations in environmental temperatures between 0^{o}C and 60^{o}C. In order to study the relationship between the characteristics of the Pre-IA conditions (60^{o}C/48 h) of PMMA dosimeters and the ambient temperatures, seventy sheets of PMMA were annealed at 60°C and stored for period of 48 h (60°C /48 h), then were kept at different temperatures of 0, 10^{o}C in a refrigerator, and at 25, 40, 50 and 60^{o}C in a thermostatically oven.

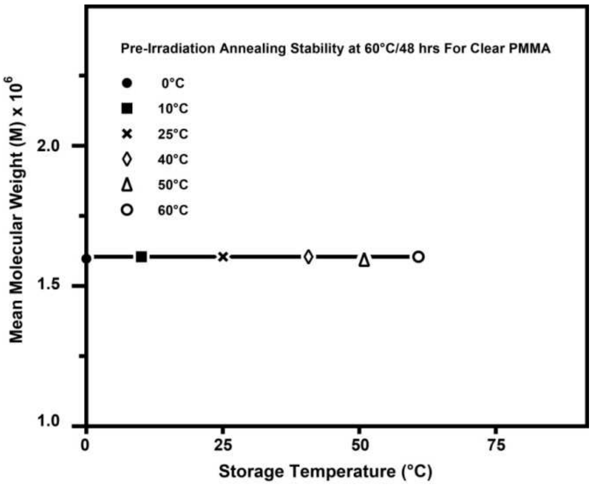

Figure 2: The Pre-Irradiation Stability of the Mean Molecular Weight as a Function of Storage Temperatures of Un-Irradiated Clear PMMA Dosimeters after Applying Pre-IA Technique at 60^{o}C/48 h

Figure 2 represents the relation between the mean molecular weight of PMMA and storage temperatures after applying Pre-IAT (60^{o}C/48 h). The results of this study shows that the pre-irradiation stability of the mean molecular weight for the PMMA dosimeters at (60^{o}C/48 h) when stored at various temperatures (0, 10, 25, 40, 50 and 60^{o}C). This result is in agreement with data published by Khan et al., (1988).

Pre- and Post-Irradiation Fading Characteristics

To study the pre-irradiation fading (Pre-IF) characteristics, sixty sheets of PMMA were stored for different periods of 1, 5, 10, 20 and 30 days after applying Pre-IAT (60^{o}C/48 h). Figure 3 shows the relation between the mean molecular weight and the storage time of the unirradiated PMMA dosimeters. The experimental results of this study show that the response of PMMA dosimeter is stable for at least 30 days. This is in agreement with the data reported by Khan et al., (1988); their results show that the response of clear PMMA dosimeter is stable for at least 15 days and that for PVC dosimeters for 30 days.

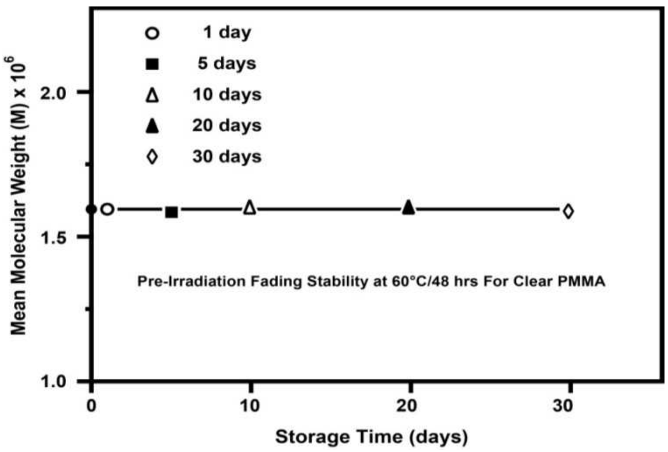

Figure 3: Relationship between the Mean Molecular Weight and Storage Time of Pre–Irradiated Clear PMMA Dosimeters after Applying Pre-IAT (60^{o}c/48h)

The stability response of clear PMMA dosimeter during post-irradiation (Post-I) has been investigated for a period up to 90 days. One hundred PMMA dosimeters were annealed at 60ºC for a period of 48 h (60^{o}C/48 h), were irradiated with test absorbed dose of 0.5 Gy from ^{90}Sr beta source, and then stored for periods of 1, 5, 10, 20, 30, 40, 60, 80 and 90 days.

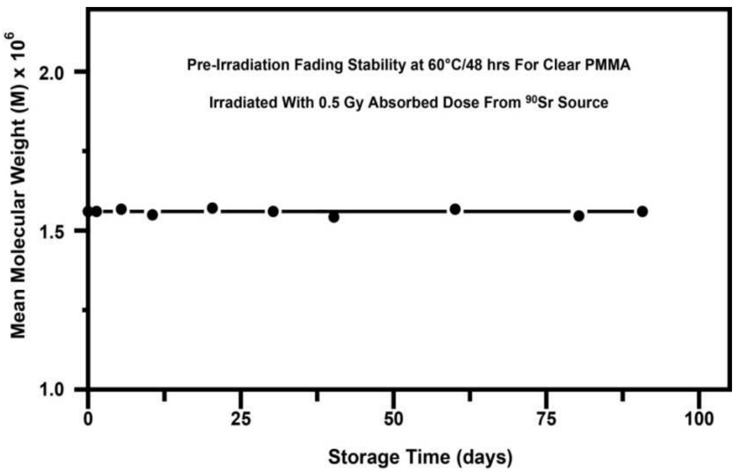

Figure 4: Post- Irradiation Fading Stability of Clear PMMA Dosimeter after Irradiated with 0.5 Gy Test Absorbed Dose from β-Particle Source as a Function of Fading Time after Applying Pre-IA (60^{o}C/48 h)

The experimental results in Figure 4 shows that at 0.5 Gy absorbed dose of beta particles there is no change in the mean molecular weight over a period up to 90 days after irradiation. The response of PMMA dosimeter investigated in the present work show post-irradiation fading stability for at least 90 days. This is due to the repair of the physical damage caused to the material during irradiation as reported by Whittaker and Watts, (2001); Khan et al., (1993); Kojima et al., (1992); Seito et al., (2009). This study indicated that the clear PMMA used as a personnel dosimeter for beta-particles shows a good stability of pre- and post-irradiation fading and is in agreement with the data reported by Devi et al, (2013); Carinou et al., (2011); Doremus and Higgins (1994).

Dose–Response

Pre-IAT (60^{o}C/48 h) have been applied to all PMMA dosimeters, and then irradiated at room temperature (25^{o}C) by beta particles from ^{90}Sr source with a dose range from 20 mGy to 10 Gy. Figure 5 illustrates the relation between the

mean molecular weight of clear PMMA and beta particle absorbed dose. Irradiated samples shows a decrease in the molecular weight with increasing absorbed dose from 20 mGy to 10 Gy, which is attributed to the main chains in macromolecular structure are cut by ionizing radiation, indicating that the polymer chains shrank in size [32] and reducing average molecular weight [1, 2, 5, 11, 12 18, 20].

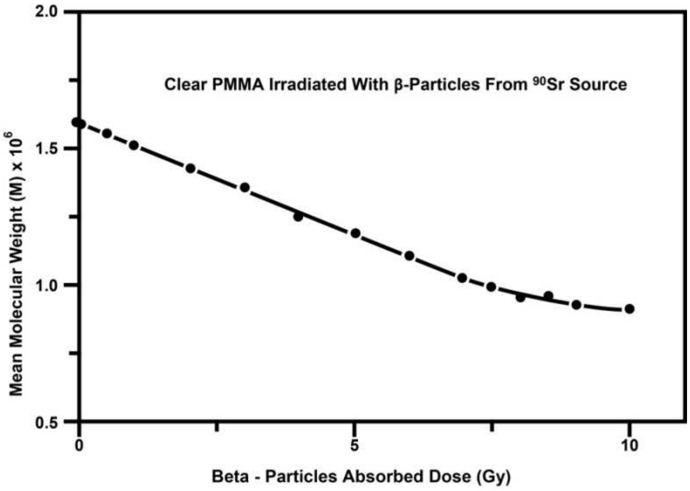

Figure 5: Changes in the Mean Molecular Weight of Clear PMMA Dosimeters as a Function of Β-Particles Absorbed Dose after Applying Pre-IAT at 60^{o}c/48 H

CONCLUSIONS

The optimum pre-irradiation annealing technique at 60^{o}C/48 h is applied to the clear PMMA personnel dosimeters in order to eliminate the effects of the variation of temperatures during winter and summer seasons and increasing the sensitivity of the PMMA personnel dosimeters. This study indicated that the clear PMMA polymers used as a personnel dosimeter for beta-particles shows a stability of pre- and post-irradiation fading for at least 90 days. Irradiated PMMA dosimeters with beta particle dose range from 20 mGy to 10 Gy undergo degradation. The results of this study show that the dosimetry characteristics of 1 mm thick clear PMMA sheets are suitable for its possible application in personnel dosimetry and can be used for a wide dose response.

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